REMEDIATION OF CONTAMINATED MARINE SEDIMENT USING BENTONITE, KAOLIN AND SAND AS CAPPING MATERIALS

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DEDICATION

This project is dedicated to the Almighty Allah, the most gracious and the most merciful, maker of all things and to my beloved family for their constant support, encouragement and love throughout this journey.

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ABSTRACT

There is a growing public concern over the issue of sediment contamination resulting from industrial, municipal wastewater, mining activities, and improper use of chemical fertilizer or pesticides. The conventional treatment of contaminated sediment is dredging, but this treatment is expensive and requires a large area of land for disposal. In situ capping of contaminated sediment is considered as a cheaper technique compared to dredging and efficient treatment technology to immobilize pollutants in sediments on site. In this technique, sediments are capped by placing a layer of inert materials like sand, clean soil, or gravel or active materials like activated carbon, zeolite, or apatite over sediments in order to reduce the risk to the aquatic environment. The objective of this study is to determine the effectiveness of using active materials; bentonite (B), kaolin (K), mixture of bentonite with kaolin (1:1) (BK) as capping materials to block the release of five heavy metals (Pb, Cr, Cu, Cd and Zn) from artificially polluted sediments. The effectiveness of B, K, and BK for preventing the leachability of the trace metals was assessed on a bench-scale laboratory experiment in glass tanks for 90 days, where 1cm thick layer of capping material and sand was placed above the contaminated sediment. The results showed that B and BK reduced the leachability of Pb, Cr, and Cu from the sediments. The results also showed that B and BK could be used as potential capping materials for the remediation of contaminated sites due to their significant entrapping of Pb, Cu, and Cr. The pollutants were released into the overlying water from the contaminated sediment in the following decreasing order; Cd > Zn > Pb > Cu > Cr. The adsorption kinetics analysis also showed that the process of adsorption was by chemisorption. This study proved that bentonite and mixture of bentonite with kaolin clays covered with sand could be used as capping materials for in situ treatment of Pb, Cu, Cr, Zn, and Cd for contaminated marine sediment.

ABSTRAK

Masyarakat secara umumnya telah menunjukkan kebimbangan terhadap pencemaran sedimen berpunca daripada pelepasan air sisa industri dan perbandaran, aktiviti perlombongan, dan penyalahgunaan baja kimia, serta racun perosak. Rawatan konvensional sedimen tercemar adalah melalui pengorekan tetapi rawatan ini adalah mahal dan memerlukan ruang yang luas untuk pelupusan. Kaedah pelapisan sedimen tercemar di tapak adalah kaedah yang murah berbanding pengorekan dan rawatan yang efisien untuk menyahaktifkan bahan cemar dalam sedimen. Kaedah ini adalah dengan menghamparkan lapisan pasir, tanah yang bersih atau bahan aktif lain seperti karbon teraktif, zeolit, atau apatit ke atas sedimen bagi mengurangkan risiko terhadap persekitaran akuatik. Objektif kajian ini adalah untuk menentukan keberkesanan penggunaan bahan aktif; bentonit (B), kaolin (K), campuran bentonit dengan kaolin (1:1) (BK) sebagai bahan lapisan bagi menghalang pelepasan lima (5) logam berat (Pb, Cr, Cu, Cd, dan Zn) daripada sedimen tercemar tiruan. Keberkesanan B, K dan juga BK dalam mencegah pelepasan logam terlarut ini diuji melalui ujian di makmal dalam tangki kaca selama 90 hari, dengan lapisan ketebalan bahan pelapis 1 cm dan pasir tersebut dihamparkan atas sedimen yang tercemar. Keputusan menunjukkan B dan BK dapat mengurangkan pelepasan logam Pb, Cr, dan Cu daripada sedimen. Keputusan juga menunjukkan bahawa B dan BK berpotensi digunakan sebagai bahan pelapik sedimen melalui keupayaannya memerangkap Pb, Cu dan Cr. Bahan pencemar dilepaskan daripada lapisan sedimen tercemar dengan turutan menurun; Cd>Zn>Pb>Cu>Cr. Analisis penyerapan kinetik juga menunjukkan proses penyerapan adalah melalui *chemisorption.* Kajian ini menunjukkan B dan BK dengan lapisan pasir boleh digunakan sebagai bahan pelapik untuk rawatan sedimen marin tercemar dengan Pb, Cu, Cr, Zn, dan Cd.

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LIST OF SYMBOLS AND ABBREVIATIONS

MNR	Monitored natural recovery
Rpm	Revolution per minute
Pb	Lead
Cd	Cadmium
Cu	Copper
Cr	Chromium
Zn	Zinc
AAS	Atomic absorption spectrometer
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
ICP-OES	Inductively coupled plasma optical emission spectrometre
Sed+B+S	Sediment + bentonite + sand
Sed+K+S	Sediment + kaolin + sand
Sed+(B+K)+S	Sediment + (bentonite + kaolin) + sand
XRF	X ray fluorescence
XRD	X ray diffraction
CEC	Cation exchange capacity
USEPA	United states environmental protection agency
EC	Electrical conductivity
B	Bentonite
K	Kaolin
BK	Bentonite + Kaolin
RAC	Risk assessment code
AAS	Atomic adsorption specstroscopy
C_o	Initial concentration of pollutant before treatment
C_t	Concentration of pollutant released into water column
T	time (number of days)
NR	No risk
LR	Low risk
MR	Medium risk

D.O	Dissolve oxygen
MMT	monmorillonite
USCS	Unified Soil Classification System
MH	Plasticity silt
Mg/L	milligram per litre
HR	High risk
VHR	very high risk
F1	Exchangeable fraction in the sequential extraction
F2	Reducible fraction in the sequential extraction
F3	Oxidisable fraction in the sequential extraction
F4	Residual fraction in the sequential extraction
ISC	Insitu capping
%	Percentage
ppm	Parts per million
UTHM	Universiti Tun Hussein Onn Malaysia

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Remediation of contaminated sediments is one of the most difficult and challenging problems in environmental engineering today. One of the primary risks associated with contaminated sediments is bioaccumulation in benthic organisms, which is a route of entry into the food chain. Thus an important goal of sediment remediation is reducing concentrations to these organisms (Lampert & Reible, 2009). Presently, due to the regular contamination of the marine sediments by the munincipal and industrial waste discharge, improper use of chemical fertilizer and pesticides, waste water irrigation and mining activities, a public concern has been aroused over the issue (Zeng *et al*., 2013a; Awad *et al.,* 2014; Teng *et al.,* 2014; Tang *et al.*, 2015). Contamination of marine sediments poses a potential threat to marine resources and human health through consumption of seafood. Marine sediments are exposed to a wide range of potential contamination by chemicals that tend to sorb to fine -grained particles. Heavy metals and organic pollutants are the two major kinds of pollutants found in sediment (Zhang *et al*., 2014; Zeng *et al.*, 2016; Zhang *et al.,* 2016b; Liu *et al*., 2016; Song *et al.*, 2017). And these are toxic contaminants, posing a great risk to the safety of food, ecological environment and human health. The problem of contamination of marine sediment is widespread in coastal water in many parts of the world. The predominant mode of heavy metal retention in sediments can be significantly influenced by the level of sediments contamination by the metal, in addition to geochemical factors and behaviours of the metal itself, metal particles frequently bind with the sediments and do not easily dissolve or migrate with water.

Several attempts have been made to find a solution to the problem of sediment contamination. So various methods and strategies have been implemented for the control and reduction of the pollution (Peng *et al*., 2009; Gerhardt *et al*., 2009; Xu *et al.*, 2012; Cheng *et al*., 2016a; Zhang *et al*., 2013a). In general, the remediation technologies can be grouped into two major strategies: in situ and ex situ remediation. In situ remediation is the treatment of the pollutant on-site, which means at the original place of contamination. This plan is targeted at the removal of the pollutant from sediment or soil without moving the sediment or soil itself. Ex-situ remediation involves excavation and treating the contaminated sediment or soil at someplace away from the site. Assessing these two strategies, in situ remediations propose several potential economic, technical, and environmental advantages (Bardos *et al*., 2000; Kuppusamy *et al*., 2016). In some instances, when considering the size of the contaminated area and the cost-effectiveness, in situ remediation is the only means of pollutant removal. Globally accepted technologies for sediment remediation generally rely on either removing the contaminated sediment, then managing it ex-situ or remediating sediment contamination in-place (in-situ). In-situ capping is an internationally recognized and accepted technology for remediating contaminated sediments and is extensively used in other countries like the USA and Norway.

For a large scale of contaminated sediment, in situ remediation brings about less disturbance to the ecosystem, the operation is comparatively easy, and the cost is more inexpensive than the ex situ treatment (Carberry and Wik, 2001; Guiwei *et al.*, 2008; Velimirovic *et al*., 2014). Capping contaminated sediments is an in situ remediation method where by a capping material is laid on top of contaminated sediments to inhibit the constant contamination of surface water and biota on top of the contaminated sediments. It involves the placing of a subaqueous cover over contaminated sediments to stabilize sediments, minimize their re-suspension and transport, and reduce dissolved contaminant transport into surface waters. It is an applicable technology which limits the release of contaminants present in the sediments by binding or adsorption, which can keep the concentration of contaminants in the overlaying water at a safe level (Gomes *et al*., 2013, Gosh *et al.*, 2011). In addition, capping treatment has less interference to the aquatic ecosystem compared to other sediment remediation technologies (Gosh *et al*., 2011,

Beckingham *et al*., 2013). Dredging and disposal are costly and may possibly bring about secondary contamination by disturbed sediments in the course of the dredging process (Nayar *et al*., 2004; Knox *et al*., 2008). Capping is assumed to be much inexpensive than dredging, and it is thought to be very effective at obstructing the diffusion of contaminants (Förstner & Apitz 2007; Perelo 2010; Sun *et al*. 2010).

Presently, materials including clay, gravel, zeolites, and activated carbon are widely applied in capping treatment methods (Jacobs & Förstner, 1999, Fadaei *et al.*, 2015). Investigation has been done on capping materials that can be used to contain inorganic contaminants, and on various capping materials used for every contaminant. Inorganic contaminants, specifically heavy metals such as Ni, Cd, Zn and Pb have been described to be successfully immobilized by phosphate minerals, including calcium phytate (Knox *et al*., 2008), apatite (Knox *et al.*, 2006; Knox *et al*., 2008) and mineral rock phosphate (Basta and McGowen 2004; Knox *et al*., 2008). More natural minerals such as limestone and zeolite were as well used as a capping material to impede the release of heavy metals (Jacobs & Förstner 1999; Eek *et al*., 2007; Knox *et al.*, 2008). In addition to heavy metals, nitrogen and phosphorus release were also stopped by calcium and zeolite minerals (Huang *et al.*, 2011; Lin *et al*., 2011; Yin *et al.*, 2013). Even though several materials have been evaluated as potential capping materials, additional research is required on low-cost and affordable material for in situ remediation of contaminated sediments.

Bentonite, a 2:1 type of aluminosillicate, is a kind of expandable clay composed primarily of montmorillonite, characterized by high permanent negative charges and large specific surface area. It has been documented that bentonite is an efficient adsorbent for some heavy metals (Karapinar and Donat, 2009; Hamidpour *et al.*, 2010; Li *et al.*, 2010).

Sediment caps incorporating certain clays, like bentonite can: (a) create a hydraulic barrier that can effectively divert flow of contaminated sediment porewaters away from migrating through the cap; (b) reduce the rate of advective transport of dissolved contaminants up into and through the cap; and (c) reduce steady-state contaminant flux through the cap more effectively than can coarser-grained materials, like sand (Reible and Lampert, 2014; USEPA, 2007; USEPA, 2013; Reible, 2008; Anchor QEA and SAO, 2014). It has been well accepted that heavy metal contaminants are retained by clayey soils through three main

mechanisms: adsorption, surface precipitation and chemical bonding Bradl, 2004; Sposito, 2008). Bentonite, as a low-cost and efficient adsorbent, has great potential in application for removing heavy metals from sediments because of its abundance, chemical and mechanical stability, high adsorptioncapability and unique structural properties (Shi *et al.*, 2011). Removal of metal ions using bentonite is based on ion exchange and adsorption mechanisms because of the relatively high cation exchange capacity (CEC) and specific surface area of bentonite (Shi *et al.*, 2011).

Similarly the removal of metal ions using kaolinite clay is also based on ion exchange and adsorption mechanisms (Tomašević *et al.*, 2014) and kaolinite has a relative low cation-exchange capacity (CEC) [3–15 meq/100 g of clay] and smaller surface area ranged from10 to 20m^2/g (Suraj *et al.*, 1998). A number of studies concerning kaolinite clay used to remove heavy metals from aqueous solution have been reported. Recently, kaolinite clay used to remove Pb (II), Cd (II) and Ni(II) in aqueous medium has been investigated through the process of adsorption under a set of variables (Gupta & Bhattaharya, 2008). Similarly, kaolinite has been used as adsorbents for the removal of Fe(III), Co(II) and Ni(II) in aqueous medium (Bhattaharya & Gupta, 2013) and for removal of Cu(II) (Bhattaharya & Gupta, 2006) and Pb(II) (Gupta & Bhattaharya, 2005). At the moment, there is not enough data about in situ capping treatments of contaminated marine sediments using bentonite and kaolin.

Although many researchers have investigated the application of reactive capping for the sediments contaminated with nutrients and organic matters, few types of research on heavy metals in the sediments have been performed. Knowledge of the application of in situ cappings for sediments contaminated with heavy metals is insufficient (Akcil, 2015). This study aims to evaluate the effectiveness of using bentonite clay, kaolin, and sand as capping materials to prevent the release of trace metals (Pb, Cd, Cr, Zn, and Cu) from the spiked heavily contaminated marine sediment (3000 ppm). To the best of our understanding, these materials have not been used as active capping material for treating contaminated marine sediment by other researchers. Although most of the previous researches were run in batch experimental condition with no flows, which has drawbacks in field usages and small number of studies were done in a flow condition, all the tests in this study were made under a flow situation using a glass tank, and to simulate ocean wave an impeller was

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